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CRISPY MIX and *FLEXI MIX* - HIGH BORON CARBIDE CONTENT, RESIN BONDED, NEUTRON SHIELDING MATERIALS

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INTRODUCTION

The most difficult regions to provide neutron shielding for a neutron scattering instrument are the inner surfaces of collimators, sample environment chambers and detector banks. In these regions some of the neutrons that enter the shielding will be partially thermalised and scattered out of the shielding. These neutrons may then find their way to the detectors and cause errors in the recorded data. This effect is more of a problem with epithermal neutrons than thermal neutrons because the neutron capture to scatter ratio increases with energy and average interaction lengths are longer at higher energies. It is therefore necessary to ensure that the neutron capture to scatter ratio of the shielding material in the critical regions is high when epithermal neutrons are present.

Boron carbide has been used in many forms for neutron shielding because of the large thermal and epithermal neutron capture cross-section of boron-10 and because it is an inert material. However boron carbide is difficult to machine so other methods of shaping it for neutron shielding have been used. Boron carbide grit can be poured into a container such as an aluminium can or a thin polyethylene bag, it can be sintered to form sheets, or it can be bonded together with a binder.

Resin bonded boron carbide tiles have been used for neutron shielding for some time. Resins have small neutron capture cross-sections and contain hydrogen which is efficient at thermalising neutrons, but if the resin content is minimised the composite material has a large neutron capture to scatter ratio for both thermal and epithermal neutrons. Professor N Watanabe of KENS Japan has developed a recipe and technique for producing a composite with a resin content of 6%-10% by weight. In attempting to repeat these results the authors have developed two forms of resin bonded carbide that have good mechanical properties, fairly uniform density and allow for simple fabrication techniques.

"Crispy mix" and "flexi mix" are mixtures of various boron carbide grits with resin that form solid composites when heat cured. Crispy mix has a rigid structure and a boron carbide content of 97% by weight. The same mixture of grit sizes can be used with a 2% resin content, but this gives some loss in mechanical strength and the resultant composite has a crumbly surface. Flexi mix is easily bent but the penalty for this flexibility is a lower boron carbide content (90%) and therefore a much higher resin content (10%).

CRISPY MIX

Most of the techniques, described below, for producing the composites were developed using the crispy mix recipe. Three grit sizes of boron carbide were used (coarse F20, medium F40 and fine F60). Various percentage combinations of the three sizes were prepared and tested and several resin binders were tried. All test samples had a small amount of fine grit, 1% to 7%, a coarse grit content of 20% to 90% with the remainder of the mix made up from medium grit and resin. The best resin of those tested was a Ciba-Geigy Araldite resin with a Thikol Chemicals hardener in the following ratios :

Epoxy resin	MY 740	100 parts
Hardener	EM 308	50 parts

Each test mix was cast into a 6 mm tile in an open mould and qualitatively tested for "strength" (can it be broken by hand?), "even density" (can light be seen through pinpricks in the tile?), "surface integrity" (does the surface crumble when rubbed with a finger?) and "surface resin" (is the surface glossy and resinous or dull and only lightly covered with resin?). The combination of grit sizes and resin that gave the lowest resin content by weight with good mechanical properties and a dull surface was :

Coarse grit	F20	38%
Medium grit	F40	56%
Fine grit	F60	3%
Resin		3%

FLEXI-MIX

To develop flexi mix the same ratio of boron caribe grit sizes were mixed with a more flexible resin. The ratio of epoxy resin to hardener and the percentage of resin in the final mix were adjusted until a 6 mm tile of flexi mix had the right combination of flexibility, integrity and low resin content. The best resin tested was made from the following ratio of Ciba-Geigy Araldite components :

Epoxy resin	CY 208	100 parts
Hardener	HY 219	25 parts
Accelerator	DY 219	2 parts

The mixture must then be oven cured at 120°C for 2 hours.

The best mix had the following ratio of boron carbide grit sizes and resin:

Coarse grit	F20	35%
Medium grit	F40	52%
Fine grit	F60	3%
Resin		10%

PREPARATION OF THE UNCURED MIX

Preparation of the two mixes is similar. The resin components are preheated to 60°C before mixing. Preheating reduces the viscosity of the resin and produces a more even distribution of resin when mixed with boron carbide grit. The boron carbide grits are dry mixed before adding hot resin and mixing. Great care must be taken to ensure that all globules of fine grit are broken up and evenly mixed with the rest of the grit and that the grit is evently wetted by the resin. All test batches were hand mixed with a steel spatula. To ensure even mixing no more than 400g of the mix were made in one batch, the grits were mixed for at least 4 minutes and the resin mixed with the grit for a further 10 minutes. A power-assisted mixer should raise the batch size limit. The raw mix has a damp appearance and no cohesive structure. It must be held in a mould and hot cured to produce the solid composite.

CRISPY MIX MOULDS

Three types of mould have been used to shape crispy mix during the curing stage: open moulds, closed moulds and hot-press moulds.

Open moulds have a base and sides made from aluminium and an open top. Shapes PTFE blocks have been inserted into some moulds to produce holes and surface profiles. The moulds are lined with thin sheets of PTFE, the mixture tamped into the mould and baked at 80°C for a time that depends on the maximum thickness of the mix (90 minutes for 60 mm and 4 hours for 50 mm). The moulds are then left to cool before removing the composite.

Closed moulds are used to produce awkward shaped structures such as thin curved pieces. The mixture is semi-cured for half the normal curing time in flat open moulds that have similar dimensions to the final shape. It is then transferred to the final mould in its green state, where it can be stretched, bent and joined surface to surface. The mix is enclosed on all sizes and returned to the oven to complete the curing.

Hot-press moulds were used to produce "high-density" flat tiles. A piece with the shape of the tile was removed from the centre of a 6 mm thick ground steel plate and the cut edge sprayed with PTFE releasing fluid. The mould was filled with crispy mix and placed between two ground steel pressure plates. To prevent the tiles sticking to the pressure plates 2 sheets of 1 mm PTFE were sandwiched between the plates and the side moulds. A pressure of 300 kg/cm^2 and a temperature of 80°C was applied for 1 hour. The moulds were then left to cool before removing the tiles.

FLEXI MIX MOULDS

So far flexi mix has only been used to cast 6 mm thick flast tiles in an open mould, but the authors see no reason why it should not be produced using the same range of techniques as crispy mix. The mixture must be baked in the mould at 120°C for 2 hours. The mould is then left to cool before removing the tile.

CRISPY MIX TESTING

Most of the testing done during the development of crispy mix has been a qualitative comparison of one tile with another and a subjective assessment of the various fabrication techniques. The major findings are outlined below :

- A resin with a little flexibility produces a more robust tile than a harder resin.

- If the resin content is reduced to 2% there is some loss of mechanical strength and the composite has a crumbly surface.

- When cast into an open mould the composite had a density of 1.45 g/cm^3 (cf 2.52 g/cm^3 for solid boron carbide). When hot pressed a density of 1.8 g/cm^3 could be achieved but the tile was difficult to remove from the mould and separate from the PTFE sheet. A hot pressed tile with a density of 1.7 g/cm^3 could be easily removed from its mould.

- While a minimum thickness of 6 mm is recommended for a robust tile, tiles have been cast with 3 mm thick by 10 mm wide lips. These lips can withstand minor knocks but break when flexed or roughly treated.

- A 6 mm thick tile was cast in an open mould and cut into five 20 mm wide by 120 mm long strips and submitted to a flextural test. The transverse modulus of rupture of the tile was 3.08 MNn m^{-2}.

FLEXI MIX TESTING

The criteria used to select a good flexi mix were that a 6 mm thick flat tile should not crack when bent through a radius of 200 mm, should not rip easily, should bend easily and have a low resin content. No quantitative tests of flexi mix properties have been made. Reducing the resin content to 6% produces a tile with a little flexibility but the tile can be easily cracked.

USAGE

Crispy mix was developed and first used to make neutron collimation irises and instrument shielding for the SNS eVS spectrometer. Open moulds have been used to produce detector shielding, flat 6 mm thick tiles to line the inside surface of the sample and detector chambers and 50 mm thick irises have been used to produce a low albedo collimation assembly. Closed moulds have been used to produce curved 6 mm thick tiles to line a flight tube that preceeds the sample chamber. Hot-press moulds have been used to produce flat 6 mm thick high density shielding tiles for regions close to the detectors. It has since found many uses on other SNS instruments and will be of great use in future SNS instruments such as SANDALS.

Flexi mix tiles are currently being used to make instrument shielding for POLARIS, the SNS polarised neutron spectrometer. The flexibility of the tiles allows them to follow the curvature of the inner walls of the instrument vacuum chamber. This saves the time that would otherwise be needed to prepare moulds for rigid crispy mix tiles, as a single mould can be used to cast standard shaped tiles which can then be cut, drilled and bent into the required shapes.

CONCULSION

While these experiments were qualitative rather than quantitative and have not exhausted the possibilities of grit ratios or fabrication techniques, a method of producing rigid neutron shielding in a variety of shapes from a 97% boron carbide/3% resin mix and flexible neutron shielding from a 90% boron carbide/10% resin mix has been developed. If a little care is used the shielding materials can be filed, drilled and cut.

Some ideas for further improvements to crispy mix include: development of a mix using a non-hydrogenous binder and introducing more flexibility into the mix without increasing the resin content. With a resin content of only 3% neutron scattering from the carbon in the boron carbide is greater than that from the resin. It may be possible to replace the boron crbide with another inert form of boron that has a lower scatter to capture ratio.

Because of their flexibility, flexi mix tiles , and presumably other moulded flexi mix structures, are easier to produce and install than crispy mix tiles but they contain three times as much resin. Where the lowest neutron capture to scatter ratio is not the sole criterion for choosing neutron shielding material, flexi mix provides a quick solution for shielding the critical areas of neutron spectrometers and diffractometers against thermal and epithermal neutrons.

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